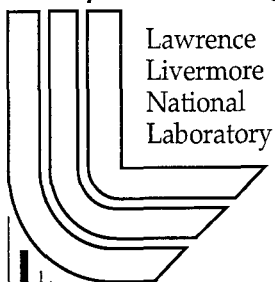# Analysis of Bulk DKDP Damage Distribution, Obscuration and Pulse Length Dependence

M. D. Feit, A. M. Rubenchik, M. Runkel

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

December 15, 2000

Analysis of bulk DKDP damage distribution, obscuration and pulse length dependence *

M.D. Feit, A.M. Rubenchik, and M. Runkel
Lawrence Livermore National Laboratory
7000 East Ave., mail stop L-477
Livermore, CA 94550

Abstract

Recent LLNL experiments reported elsewhere at this conference explored the pulselength dependence of 351 nm bulk damage incidence in DKDP. The results found are consistent, in part, with a model in which a distribution of small bulk initiators is assumed to exist in the crystal and the damage threshold is determined by reaching a critical temperature. The observed pulse length dependence can be explained as being set by the most probable defect capable of causing damage at a given pulselength. Analysis of obscuration in side illuminated images of the damaged region yields estimates of the damage site distributions that are in reasonable agreement with the distributions experimentally directly estimated

Keywords: laser damage, KDP, obscuration, pulselength scaling

INTRODUCTION

The frequency conversion crystals of KDP (doubler) and deuterated KDP (DKDP, tripler) used in high power lasers such as the National Ignition Facility (NIF) are susceptible to bulk laser induced damage. This damage, which does not grow catastrophically upon re-irradiation at the same fluence, nevertheless must be such as to not obscure a significant amount of the propagated laser beam. LLNL has an extensive damage test effort to characterize this damage which manifests as a multitude of small "pinpoint" bulk damage sites and to relate this damage to obscuration.

Since damage tests are carried out on several lasers at different pulselengths, it is necessary to know how to relate damage behavior from one pulselength to another. Additionally, pulselength dependence gives important information about the underlying physical mechanisms, which set the threshold. The theoretical treatment in this paper together with a companion experimental paper in these proceedings give an account of observations of pulselength dependence of number and size of pinpoint damage sites in DKDP in the UV (351 nm) and the resultant beam obscuration.

The following sections develop the model of a small absorbing "particle" as damage initiator. It is assumed that heating the particle to a threshold temperature determines initiation of damage. The novel aspect of this treatment is that absorbers of different sizes are most efficiently heated by pulses of different lengths. If a distribution in size of very small initiators exists, this model predicts a dependence on pulselength τ of damage threshold of form τ^m where $m \leq 0.5$. This prediction is in accord with the present observations and other results available in the literature. Combined with an independent observation of total beam obscuration, the model allows prediction of the smallest scale pinpoints contributing to damage.

LASER ABSORPTION BY SMALL PARTICLES

We consider spherical particles for simplicity. Such particles may be much smaller than an optical wavelength and thus very difficult to detect directly. Absorption of laser energy and the resulting temperature rise in the surrounding material, and, therefore, the incidence of damage, is expected to be a function of particle size and absorption coefficient as well as laser fluence and pulse duration. In principle, comparison of the damage density at different laser intensities and wavelengths can yield information on the initial particle size distribution. We considered both small "black", i.e. highly

absorbing, and larger "grey", i.e. moderately absorbing particles. We calculated the particle absorption cross sections using Mie theory [1] for particles of arbitrary size or absorptivity using a code based on the algorithm of Wiscombe [2].

The absorption efficiency α, i.e. the fraction of light incident on the geometric cross section that is absorbed is defined by $\alpha = \sigma_a/\pi a^2$ where a is the particle radius and σ_a is the absorption cross section. The important parameter is the ratio of particle size to absorption length. Starting from a very small size 2a, α will initially grow at least linearly with a, and then tend to oscillate around a saturated value. Because of diffraction, the geometric absorption fraction can be larger than unity as shown by the results plotted in Fig. [1]. The transparent substrate has index 1.4765 and the absorbing particle was assumed to have refractive index 2.0. Results for values of the imaginary part of the particle's index ranging from 0.01 to 0.2 are shown (typical values for strong absorbers are 0.2 for ceria and 0.25 for SiO). Contours of the absorption efficiency as a function of particle size and imaginary part of the particle index are shown in Fig. (2).

HEATING BY ABSORBING SMALL PARTICLES

The temperature distribution around an absorbing spherical particle subject to a constant laser flux will follow the heat equation:

$$\frac{\partial T}{\partial t} = D\, \nabla^2 T\ + Q(r)$$

(4)

where D is the thermal diffusivity and the spatially dependent but time independent source term Q can be taken in the form $Q(r) = \alpha\, \pi a2\, I\, \exp(2\, K\, k[r-a])/\rho\, C$ inside the particle and $Q=0$ outside the particle. Here I is the laser intensity, α is the absorption efficiency as before, K is the wavenumber, $K= 2\pi/\lambda$, k is the imaginary part of the index, a the particle radius, ρ the mass density, and C is the specific heat.

We motivate the numerical solution by considering first a simple approximation for a perfect conductor that is more amenable to intuition. Suppose that at time t, energy is being deposited in a spherical particle of radius a. The temperature inside the particle is uniform due to its perfect conductivity. Furthermore, because of conduction the energy deposited extends a thermal distance $\sqrt{4Dt}$ into the surrounding medium. Then, the particle temperature will be described by an equation of form

$$\rho C \frac{dT}{dt} = \frac{\alpha\, I\, \frac{4}{3}\, \pi a^3}{\frac{4}{3}\, \pi a^3\ + 4\pi a^2 \sqrt{4Dt}}$$

$$= \frac{\alpha\, I}{1\ + 6\, \sqrt{Dt\,/\,a^2}}$$

(5)

where α is an inverse absorption length and D is the thermal diffusivity. Integrating this equation over a laser pulse of duration t predicts a final temperature of

$$T(t) = \alpha\, I\, a^2\, (6\, \sqrt{t} - \log[1+6\sqrt{t}])\, /\, (18\, K)$$

(6)

where $\tau = Dt/a^2$ and K is the thermal conductivity of the substrate. The interesting part of this result is that the temperature scales very nearly as $I\sqrt{t}$ or $F/\sqrt{t}$ with F being the fluence. Thus, if a damage threshold is determined by a critical temperature, one would expect the fluence threshold to scale slightly slower than as $\sqrt{t}$.

An exact solution[3] of the heat equation, valid up to intermediate times describes a spherical perfect heat conductor of radius a containing a heat source. The source particle is embedded in an initially cold material of finite conductivity and radius b. The temperature at r=b is maintained at 0. This solution is given by

$$T(r,t) = QQ$$

$$\left[\frac{1}{r} - \frac{1}{b} - \frac{4\gamma}{r\,a}\sum_{n=1}^{\infty} \frac{\sin\left(\beta_n(b-a)\right)\sin\left(\beta_n(b-r)\right)\exp\left(-\beta_n^2 Dt\right)}{\beta_n\left(2\gamma(b-a)\beta_n + 4\beta_n a \sin^2\left(\beta_n(b-a)\right) - \gamma\sin\left(2\beta_n(b-a)\right)\right)}\right]$$

(7)

where $QQ = \alpha\, I\, \pi a^2/Ka$, α=absorption efficiency, I=laser intensity, K=thermal conductivity of substrate, $\gamma = 4\pi a^3 \rho C/M'C'$, ρ and C are the density and specific heat of the substrate, M' and C' are the mass and specific heat of the particle, and β_n is a solution to the transcendental equation

$$\gamma\,\beta_n a\,\cos\left(\beta_n(b-a)\right) = \left(\beta_n^2 a^2 - \gamma\right)\sin\left(\beta_n(b-a)\right)$$

(8)

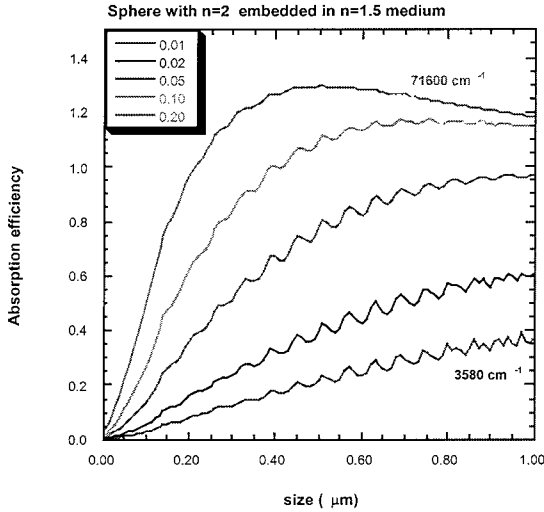An appealing intuitive approximate solution to Eq.(4) is given in ref.[4].

Fig. 1: Fraction of laser energy incident on geometric cross section that is absorbed by a particle whose diameter is given on the x-axis. Curves correspond to values for the imaginary part of the particle refractive index yielding absorption coefficients from 3580 to 71600 cm^{-1}.
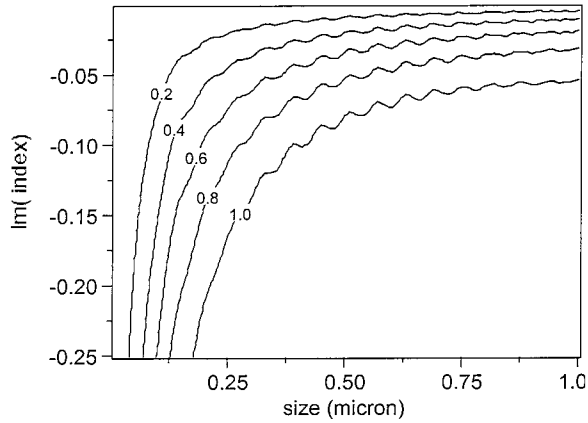
Fig. 2: Contours of absorption efficiency for 1 μm and smaller particles with varying degrees of absorptivity.

The heat equation introduces another length scale, the diffusion length $\sqrt{D\tau}$. The absorbing particle may have thermal properties similar to or much different from the surrounding material. We considered two limiting cases to bracket realistic behavior. The first assumed the particle has the same thermal conductivity as the host material. The second assumed that the absorbing particle has much higher thermal conductivity than the surrounding material. These extremes do not lead to qualitatively different behavior.

Even when the absorber and matrix have the same conductivity, two different histories occur depending on whether the particle size is large or small compared to the thermal diffusion length. For particles small compared to the diffusion length, a steady state temperature distribution is reached in the particle at which the energy gained through absorption is just balanced by the heat flux out of the particle. At long times, the temperature outside the particle decreases as $1/r$. For particles large compared to the diffusion length, nearly all of the deposited energy is used to raise the particle temperature. Said differently, for particles of a given size exposed to laser radiation, the temperature first increases linearly with time, and then tends to
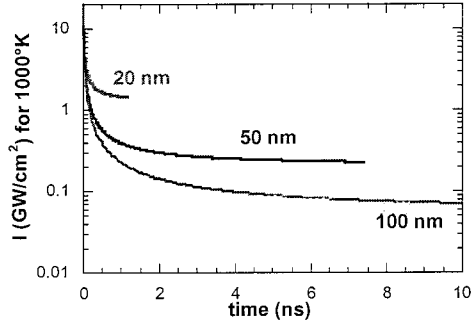
Fig. 3:Intensity in GW/cm2 required to heat particles of size 20,50 and 100 nm by 1000°K assuming Im(η)=0.02
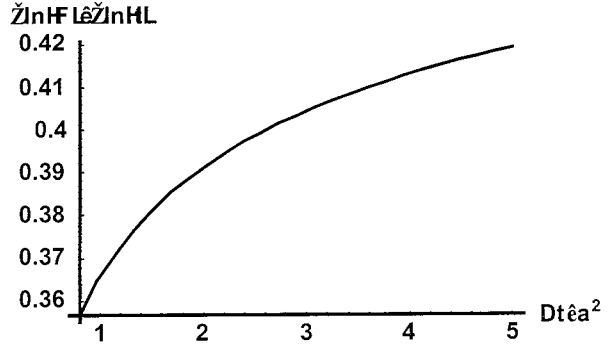

Fig. 4: Effective powerlaw exponent, i.e. logarithmic derivative of threshold fluence with respect to normalized pulse duration.

level off once heat conduction becomes a significant energy loss mechanism. This is shown in Fig.(3) where we see that larger particles continue to heat longer and therefore require a smaller laser intensity to reach a given temperature.

With this scaling and the absorption efficiencies found earlier, we can estimate the laser intensities needed to heat particles in a distribution.

EFFECT OF SIZE DISTRIBUTION

Suppose there is a distribution in size of absorbing sites with a higher density of small sites than of large sites. As the laser pulselength is varied, it is clear from the above that the most efficient heating to a given temperature occurs for particles of size about equal to the corresponding thermal diffusion length. That is different sized absorbers in the distribution determine the threshold for different pulselengths. The net result, as shown in Fig.(4), is a damage initiation threshold that scales as a power of pulselength equal to 0.35-0.45 consistent with the experimental observations reported in the next section.

COMPARISON WITH OBSERVATIONS

Recent observations[5] of the pulselength dependence of KDP bulk damage incidence have been made. These experiments revealed that the threshold fluence, number of "pinpoint scatterers" and size of defects all varied with pulselength. Some recent observations suggest damaged regions in bulk may be anisotropic. There is thus some uncertainty in interpreting the previous data on number and size of defect regions since results may vary with angle of observation. We concentrate here on the observed threshold fluences as a function of pulse duration.

The model calculations above indicate that if the initiating particles are very small compared to a thermal diffusion length (small compared to 200 nm), the damage threshold should depend only on laser intensity. Thus, the threshold damage fluence should vary linearly with pulse duration. Alternatively, if the initiators are large compared to the thermal diffusion length, damage incidence depends on the total energy deposited and the threshold fluence should be independent of pulse duration. In the intermediate case where initiators are about the same size as the thermal diffusion length, the damage threshold should vary as an intermediate power of pulse duration near 1/2.

When the number of detected scatterers due to DKDP bulk damage are plotted vs. laser fluence, the three pulse durations yield distinct curves. Because of some uncertainty in the normalization of the total number of sites counted, we normalize the curves to unity to more easily see differences in shape. Even more dissimilar curves are obtained if the results are plotted vs. laser intensity. However, the three curves (except for normalization) tend to overlay each other when plotted against fluence divided by a power of the pulse duration slightly less than 0.5 as shown in Fig.(5). The best fit is for a power of 0.35, which is consistent with the results of calculation shown in Fig.(4).

The size distribution of sites at two pulselengths is shown in Fig.(5). In both cases, the number of sites scales as the inverse cube of size. Assuming lower and upper size cutoffs, this implies a total obscuration that depends on these cutoffs and the total number of sites. Combined with an independent measurement of obscuration, this suffices to fix the minimum site

size. Using the obscuration determined by photometer measurements reported in ref[5], this yields a minimum size of 1-2 μm, consistent with observation.
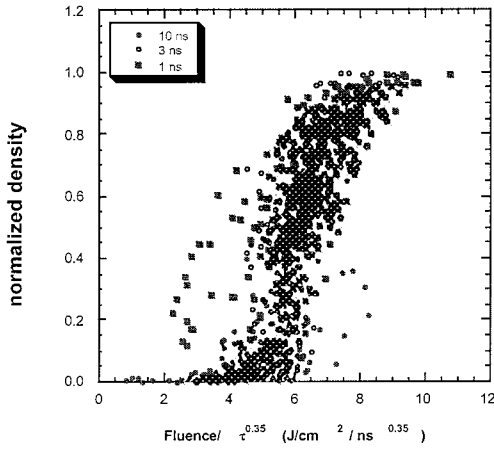

Fig. 5: Normalized observed DKDP damage incidence plotted as a function of fluence/ $t^{0.35}$ Curves for the three pulse durations are now quite similar
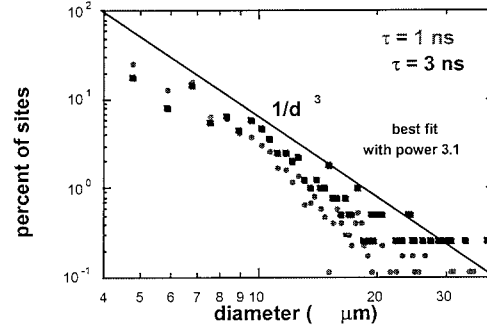
Fig. 6: Distribution by size of bulk damage sites contributing to obscuration at two pulselengths. Combined with independent measurement of obscuration, this allows determination of lower size cutoff value.

CONCLUSION

We have developed theoretical models of particle absorption and heating which indicate that particles a few hundred nm in size of material with imaginary refractive index as small as 1% that of ceria can cause heating by 1000°K.

A general picture of heating of "small" and "large" (compared to thermal diffusion length) particles in a distribution of particles with many more small particles than large particles suggests that thresholds for a critical temperature would be set by particles of about the thermal diffusion length size and that fluence thresholds would then vary approximately as the square root of the pulse duration. This later expectation is borne out by LLNL observations of pulselength dependence of bulk DKDP damage. The heating effects described here thus offer a realistic possible explanation of the generally observed $\sqrt{t}$ dependence noted for damage in many materials including fused silica[6].

References

[1] *Light scattering by small particles*, H.C. van de Hulst, John Wiley and Sons, NY (1957)

[2] "Improved Mie scattering algorithms", W.J. Wiscombe, Appl. Opt. **19**, 1505 (May 1, 1980)

[3] *Conduction of heat in solids*, H.S. Carslaw and J.C. Jaeger, Clarendon Press, Oxford (1959)

[4] "Pulsed laser induced damage of an optical material with spherical inclusion", M.R. Lange, J.K. McIver, A.H. Guenther, and T.W. Walker, Damage in Laser Materials:1982, Nat. Bur. Stand. (U.S.) Spec. Publ. **669** , 380-6, (1984)

[5] "Results of pulselength scaling experiments on rapid-growth DKDP triplers using the Optical Sciences Laser at 351 nm", M. Runkel, A. Burnham, D. Milam, W. Sell, M.D. Feit, A.M. Rubenchik, elsewhere in this proceedings

[6] D. Milam, "1064 NM Laser Damage Threshold of Polished Glass Surfaces as a Function of the Pulse Duration and Surface Roughness," Laser Induced Damage in Optical Materials: 1978, A. J. Glass and A. H. Guenther Eds., NBS Spec. Pub. **541**, 164 (1978)